1.0

2.8 | 2.5

3.2 | 2.2

3.6

4.0 | 2.0

1.1

1.8

1.25 | 1.4

MICROCOPY RESOLUTION TEST CHART

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COMPTON SCATTERING STUDIES

OF

METALS AND HYDRIDES

WITH

412 KeV γ-RADIATION

by

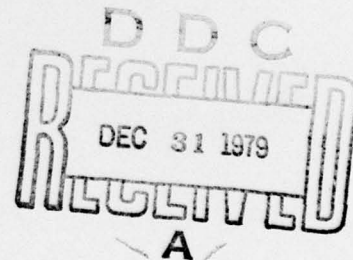Malcolm J. Cooper

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A 412 KeV Compton spectrometer has been designed at Warwick, and commissioned at the SRC Rutherford Laboratory. The performance of the system was highly satisfactory, but routine operation of the equipment has been hampered by deterioration in the resolution of the detector, and delays have occurred as a result of faults in the computerised data acquisition system. The Compton profile of vanadium has been measured, and is found to be in disagreement with theory, the anisotropies being smaller than predicted.

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ABSTRACT

A 412 KeV Compton spectrometer has been designed at Warwick, and commissioned at the SRC Rutherford Laboratory. The performance of the system was highly satisfactory, but routine operation of the equipment has been hampered by deterioration in the resolution of the detector, and delays have occurred as a result of faults in the computerised data acquisition system.

The Compton profile of vanadium has been measured, and is found to be in disagreement with theory, the anisotropies being smaller than predicted.

In related investigations evidence for the failure of the impulse approximation has been found, and the viability of obtaining cohesive energies from Compton profiles has been assessed.

KEYWORDS: Compton Profile, Compton spectrometer, vanadium, cohesive
energies.

CONTENTS

Progress at the Rutherford Laboratory during the second year was in fact marred by hardware and software faults on the PDP/8 data acquisition system provided by the Rutherford Laboratory; the delays were exacerbated by the deterioration of the Ge(Li) solid state detector. Both these problems have now been solved or circumvented by SRC-funded purchases, but progress during the second year of the grant was minimal. Indeed the 412 KeV data on vanadium, briefly reported in section 3 was obtained by Dr. Holt during a visit to the Hahn-Meitner Institute in Berlin. Vanadium will be studied further in an attempt to reconstruct the 3-D momentum distribution in the metal, and to correlate those results with X-ray diffraction and positron annihilation data. Unlike niobium where the theoretical description of the charge density is in reasonable accord with experiment, vanadium is poorly described by current theory. Hopefully further experiments will provide the stimulus for more detailed calculations.

In view of the lull in work with the 412 KeV source related problems were tackled by the investigator and Professor DuBard using the 60 KeV ^{241}Am source at Warwick University. The principal of these was the discovery of a small but significant asymmetry in the Compton profile of aluminium (and heavier materials) when 60 KeV radiation is used. This failure of the impulse approximation has serious implications for Compton scattering work at these energies. This work, described in section 4, has recently been published in the Philosophical Magazine (vol. 39B, p541, 1979).

Finally the problems associated with the derivation of cohesive energies from Compton data have been investigated. For materials with a low vapour pressure, or a very high melting point the derivation of binding energies in this way may constitute the only practicable and non-destructive method possible. The preliminary survey reproduced in section 5 forms the basis of a paper accepted for publication in the Philosophical Magazine.

Section 2 : THE 412 KeV SPECTROMETER

§ INTRODUCTION

Electron momentum density can be studied in a Compton scattering experiment because the Compton scattered radiation is, in effect, Doppler broadened by virtue of the electron's motion; to be specific the Compton profile is given, with certain simplifying assumptions, by the projection of the electron momentum distribution onto the scattering vector. A general review of the subject has been given by Cooper (1977) while a detailed account is presented in a book edited by Williams (1977).

Within this decade the use of crystallographic X-ray sources for Compton scattering studies, in conjunction with crystal spectrometers for the energy analysis, has been superceded by higher energy γ-ray studies, and the spectrometers have been replaced by energy sensitive germanium detectors. There were at least two reasons for this change. Firstly on a practical level in all but the lightest elements photoelectric absorption

is prohibitively high at X-ray wavelengths and the weak Compton scattering suffers a drastic reduction in intensity. Secondly a more fundamental objection arises at low energy because the 'Impulse Approximation' necessary for the interpretation of the scattering cross-section in terms of electron momentum, as outlined above, is invalid if the energy of the recoiling electron is comparable with its binding energy. Again for elements heavier than, say, argon this requirement is clearly violated when conventional X-ray sources are used.

Ideally a γ-ray source for Compton scattering would have a high specific activity, a long half-life, monochromatic γ-ray emission preferably in the range 100 KeV - 400 KeV, and it should be cheap to produce! However these requirements cannot all be met simultaneously, indeed the first two are mutually exclusive. In practice γ-ray work has centred on the isotopes ^{241}Am with E_γ = 59.5 KeV, $T_{\frac{1}{2}}$ = 458 yr. (see, eg. Cooper, Holt, and DuBard 1978), ^{123m}Te with E_γ = 159 KeV, $T_{\frac{1}{2}}$ = 107 days (see e.g. Eisenberger and Reed, 1972). In addition ^{137}Cs with E_γ = 662 KeV and $T_{\frac{1}{2}}$ = 30 yr., (see, e.g. Cooper, Pattison and Schneider, 1976) have both been used. A discussion of the advantages and disadvantages of various radiation sources (X-radiation, γ-radiation, synchrotron radiation) has been given by Cooper, 1979; this paper is pecifically concerned with the construction of a new spectrometer for the use of the short-lived gold isotope ^{198}Au.

When gold is bombarded with neutrons, the following reactions take place

$$^{197}Au_{(n,\gamma)} \quad ^{198}Au_{T_{\frac{1}{2}}=2.7 \text{ days}} \quad ^{198}Hg + \gamma \quad (412 \text{ KeV})$$

$$^{198}Au_{(n,\gamma)} \quad ^{199}Au_{T_{\frac{1}{2}}=3.2 \text{ days}} \quad ^{199}Hg + \gamma \quad \begin{cases} 208 \text{ KeV} \\ 158 \text{ KeV} \end{cases}$$
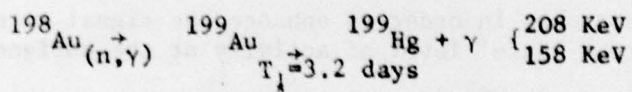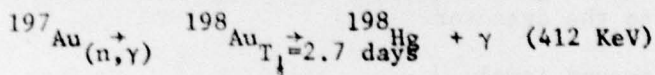
The use of the dominant 412 KeV line for diffractometry was pioneered by Schneider (1974 a, b); its particular advantage being that extinction-free diffraction measurements can be made in transmission on thick crystals. The specific activity of gold is high so that 100 Curie sources with small dimensions ($\sim$ 8 mm^3) can be produced in neutron fluxes $\sim 10^{12}$/mm^2/sec. In view of the short half-life the gold source can only be used on a regular basis in the vicinity of a reactor where a programme of activations is possible.

The first γ-diffractometer, built at the Institut Laue-Langevin (ILL) in Grenoble, used gold foil sources irradiated at the nearby SILOE reactor of the Centre d'Etudes Nucleaire de Grenoble. It was used for several Compton scattering experiments on an ocassional basis (see, e.g. Cooper, Pattison, and Schneider, 1976), but because it had been designed primarily as a γ-ray diffractometer both signal and signal to noise ratios were rather poor. It was therefore decided to design and construct a new diffractometer/

Compton Spectrometer which would be sited at the Rutherford Laboratory in the United Kingdom, using the adjacent neutron reactors at AERE, Harwell for the irradiations. The Compton spectrometer has been designed to give an order of magnitude improvement over the previous spectrometer at ILL. Another instrument recently constructed at the Hahn Meitner Institute (Pattison and Schneider, in press) gives a similar performance to the one described here, although the design is rather different.

§ DESIGN CRITERIA

Given the choice of ^{198}Au as the radiation source the design criteria can be summarised by the following points.

(a) To make the scattering angle as large as possible.

There are two reasons for this, first the ratio of the width of the Compton line to that of the detector resolution function increases with angle, and second the broadening of the Compton profile due to the finite beam divergence decreases rapidly at high angles. The latter is the dominant consideration, and taken together the two effects dictate that, in this energy range, Compton scattering measurements are made at scattering angle in excess of 150°, with beam divergences below $\pm 2^{\circ}$.

(b) To minimise the lengths of the incident and scattered beam paths.

Given that the scattering is weak this is an obvious requirement for increasing the flux into the detector.

(c) To reduce the background level.

This is necessary not only in order to enhance the signal to noise ratio, but also to ensure a 'safe' level of activity at the surface of the source block.

(d) To minimise the influence of γ-radiation from the ^{199}Au isotope which is produced from ^{198}Au under neutron bombardment.

This point was not fully appreciated in the previous 412 KeV studies; although it was noted that the 158 KeV line from ^{199}Au lies in the middle of the spectrum of Compton scattered radiation, elastic scattering of this weak line ($\sim 2\%$ of the intensity of the 412 KeV line) was considered negligible. Indeed it is for light element scatterers ($Z < 40$, say), but for heavier materials, with larger elastic scattering cross-sections, such an additional line complicates the analysis of the spectrum.

In practice it is necessary to strike a balance between these various objectives. In the ILL equipment, for example, bulky lead shielding had been used, and this in turn meant long beam paths ($\sim 0.5 - 1.0$ m) at high angles of scattering, and hence low signal levels. Even so signal to noise ratios were modest (50:1) and surface activity was an unhealthy 100-200 m rad with a 'fresh' 70 Curie source. The shielding problem is exacerbated by the presence of small amounts of very penetrating γ-radiation from ^{198}Au,

at 675 KeV (0.6%) and 1090 KeV (0.2%). Lead affords poor protection at these energies and the possibility of making the source block out of a more dense, high Z, material was explored. Although depleted uranium and gold were considered (albeit briefly!) a commercially available tungsten alloy - 'Heavy Alloy' - manufactured by Osram - GEC was chosen. This material which is produced by sintering, and can be machined in much the same fashion as stainless steel, has a density of 16.8×10^3 kgm^{-3}. By way of example at 8 cm thickness heavy alloy absorbs ten times as much radiation from ^{198}Au than lead.

In order to minimise costs the central part of the source block alone was made from heavy alloy, the outer casing being lead; even so the raw material cost, at 1977 prices, was close to £2,000.

§ CONSTRUCTION

(a) General considerations

The overall outline of the spectrometer is shown in figure 2.1; γ-radiation passes through a lead collimator into the sample chamber, and the radiation which is Compton-scattered through 167° is recorded by the lithium-drifted germanium solid state detector positioned behind the source block. The collimators and the sample chamber are sealed with mylar foil windows and evacuated to eliminate air scattering.

Given the choice of shielding material, and having determined the angle of scattering, the scale of the equipment is governed by the minimum allowable distance between the source and the scattered beam collimator. Experience with the ILL spectrometer had indicated that radiation leakage in this direction contributed disproportionately to the background in the experiment. Clearly the most efficient absorber, tungsten alloy, must be used to ensure the optimum compromise between high absorption and minimum dimensions. The source block therefore consisted of a 160 mm x 160 mm x 80 mm block of tungsten alloy with five faces clad with lead and the sixth face, adjacent to the collimator clad with tungsten alloy, to give a minimum path through that material of 70 mm. The collimator itself was constructed from a large cylinder with the beam path off-centre. In this way the effect of scattering from the walls is minimised, again reducing the background at the detector.

The level of surface activity was calculated to be less than 10 mrad hr^{-1} for the proposed dimensions, with a 100 Curie source installed. In practice the highest figure recorded to date is 5 mrad hr^{-1}.

In the study of very heavy, and hence heavily absorbing materials, signal strength may become a problem, and with this in mind a second form of the equipment has been produced, with shorter collimators. The scattering angle of 167° is maintained, but two tungsten alloy plates, each 10 mm thick, must be removed. Thus the intensity can be doubled, but at the expense of some deterioration in the signal to noise ratio.

Finally in this section it should be noted that the scattering chamber has an exit port in line with the incident beam direction so that, when Compton measurements are not being made, a simple γ-diffractometer can be used. That instrument will be described elsewhere.

(b) The Source Block

The principal features of the source block are evident from the vertical section shown in figure 2.2, the overall dimensions being derived from the shielding and safety considerations discussed above. The blocks of tungsten and lead are sandwiched between 10 mm thick steel plates, and lifting hooks are attached to the top plate so that the 127 kg block can be moved.

The shutter mechanism simply consists of a rotateable cylinder of tungsten alloy, with a transverse beam hole. A sprung pin in the base ensures that the window can only be opened when the block is in place in the spectrometer, and a microswitch attached to the window mechanism is connected to a warning sign. The source pin cannot, of course, be withdrawn by accident.

Contrary to first expectations it is important not to surround the source too closely with absorbing material since this would ensure that a significant amount of Compton scattered radiation would find its way down the incident beam collimator. The monochromatic 412 KeV line would then appear with an extensive low energy tail. In order to minimize this effect a hole, 20 mm diameter and 10 mm deep was drilled out behind the source (see figure 2).

(c) The Source Holder

This was designed in consultation with the Isotope Production Unit at AERE, Harwell: it was also made from tungsten alloy to ensure adequate shielding in the upward direction. The gold foil source (initially 6.5 mm x 6.5 mm x 0.25 mm) is sandwiched in a graphite rod 27 mm x 6.5 mm diameter and irradiated in DIDO. The graphite rod is then clipped onto the bottom of the tungsten alloy pin, and located and retained with the help of a spring-loaded bayonet cap. The source assembly is shown in figure 2.3; there are two positions of the pin corresponding to the foil being 'face-on' (for Compton scattering), or 'edge-on' (for γ-diffractometry). The holder is constructed so that it can be assembled and disassembled by remote handling in a 'hot' cell.

(d) Irradiation Strategy

In γ-ray diffractometry, where for example a well-defined line source (0.2 mm x 10 mm) may be used, the useful volume of irradiated materials is also limited by self-absorption- there is little point in making such a 412 KeV source more than three or four millimetres thick, and consequently these sources tend to have an upper limit to their activity of around 100 Curies. In a Compton scattering experiment, however, the source can have a larger cross-section since beam divergences are measured in degrees rather than minutes. Thus the same foil could be viewed 'face-on' with a consequent increase in the intensity of emission of the 412 KeV radiation. Unfortunately this effect is even more marked for the lower energy lines of ^{199}Au, and as indicated earlier this can cause additional complications in Compton scattering studies of heavy materials.

However the neutron cross-sections for the production of ^{198}Au and ^{199}Au are different, and so their ratio can be altered by varying the irradiation time. Figure 2.4 shows the activity induced in a 10 mm^3 sample of gold irradiated in DIDO. For example after a 48 hour irradiation the activity of ^{198}Au is 90 Curies and of ^{199}Au, 30 Curies. In fact the first trial experiment was carried out with a source of this composition, irradiated to saturation. With a tin sample the 158 KeV elastically scattered line of ^{199}Au was visible near the peak of the Compton scattered 412 KeV line of ^{198}Au. (At 167° the Compton peak is centred at 159 KeV).

In order to maximise the relative intensity of the 412 KeV line it is obviously preferable to irradiate a gold foil twice as thick for half as long. In that case the intensities of the lines are ∿ 120 Curie and ∿ 20 Curie respectively, which is a much more favourable ratio. Subsequent irradiations have in fact followed this prescription, two foils being sandwiched together in the same graphite holder. A further increase in thickness will probably be possible following minor modifications to the holder.

§ SUMMARY

To date several irradiations have been carried out without any problems with the irradiation or handling procedure, although data collection has been hampered by faults in the detector and the software of the data acquisition system. Various figures of merit are collected together in the accompanying table where a comparison with old ILL spectrometer and the new instrument at the Hahn-Meitner Institute, is also made.

The current improvement over the ILL instrument is readily evident in the signal levels (the figures quoted for aluminium are directly comparable) and especially in the signal to noise ratios (these are defined as the ratio of the count rate at the peak of the Compton profile, to the count rate at the same energy, but with the sample removed).

Such high figures do have a practical importance; high intensities are necessary if experiments are to be finished in one or two half-lives, and high signal to noise ratios are necessary if the tails of the profile are also to be measured accurately. The low level of surface activity would certainly allow an increase in the source activity of say a further factor of two, and this would be best achieved, as discussed in the previous section, by increasing the volume of the source.

In conclusion the Compton spectrometer has achieved the designed performance, and there is every indication that further improvements can be made. Exploratory measurements at ILL, and new measurements at the Hahn-Meitner Institute have indicated that Compton scattering studies of heavy metals, and heavy metals hydrides will be particularly fruitful. Such measurements are well within the scope of this instrument.

	Grenoble Warwick	Rutherford-Warwick	Hahn-Meitner Institut
Activity (Ci)	70	120	100 - 150
Source size (mm)	foil 0.2 x 10 x 4	foil 6.5x6.5x0.25	cylinder 2 mm diam.
Scattering angles	167° 169° 171°	167°	165°
Beam divergence (FWHM)	$\pm 0.7°$	long $\pm 0.87°$ short $\pm 0.92°$	$\pm 0.6°$
Source-sample distance (cm)	50	long 47.6 short 36.3	35
Sample-detector distance (cm)	100	long 59.4 short 48.4	65
Collimator diameter (cm)	1.6	1.12	1.6 - 0.9
Signal-noise ratio	Al Ge } 50:1	Al 1000:1 Sn 250:1 (short x .8)	Al Ge > 1000:1
Compton intensity cts/50eV/hr/mm Sample thickness	Al 56	Al 457 Sn 267 (short x 2)	Al 369 (+) Au 217
Surface activity	150 mrad hr^{-1}	5 mrad hr^{-1}	~ 2m rad hr^{-1}

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Pattison, P., Cooper, M. J., Holt, R. S., Schneider, J. R., and Stump, N., 1977, Anisotropy in the Compton profiles of Nb and NbH$_{0.76}$. Zeits. Phys. B27, 205.

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COMPTON SPECTROMETER

heavy
alloy

lead
shielding

sample chamber

solid state
detector

SCALE cms.

0   10   20
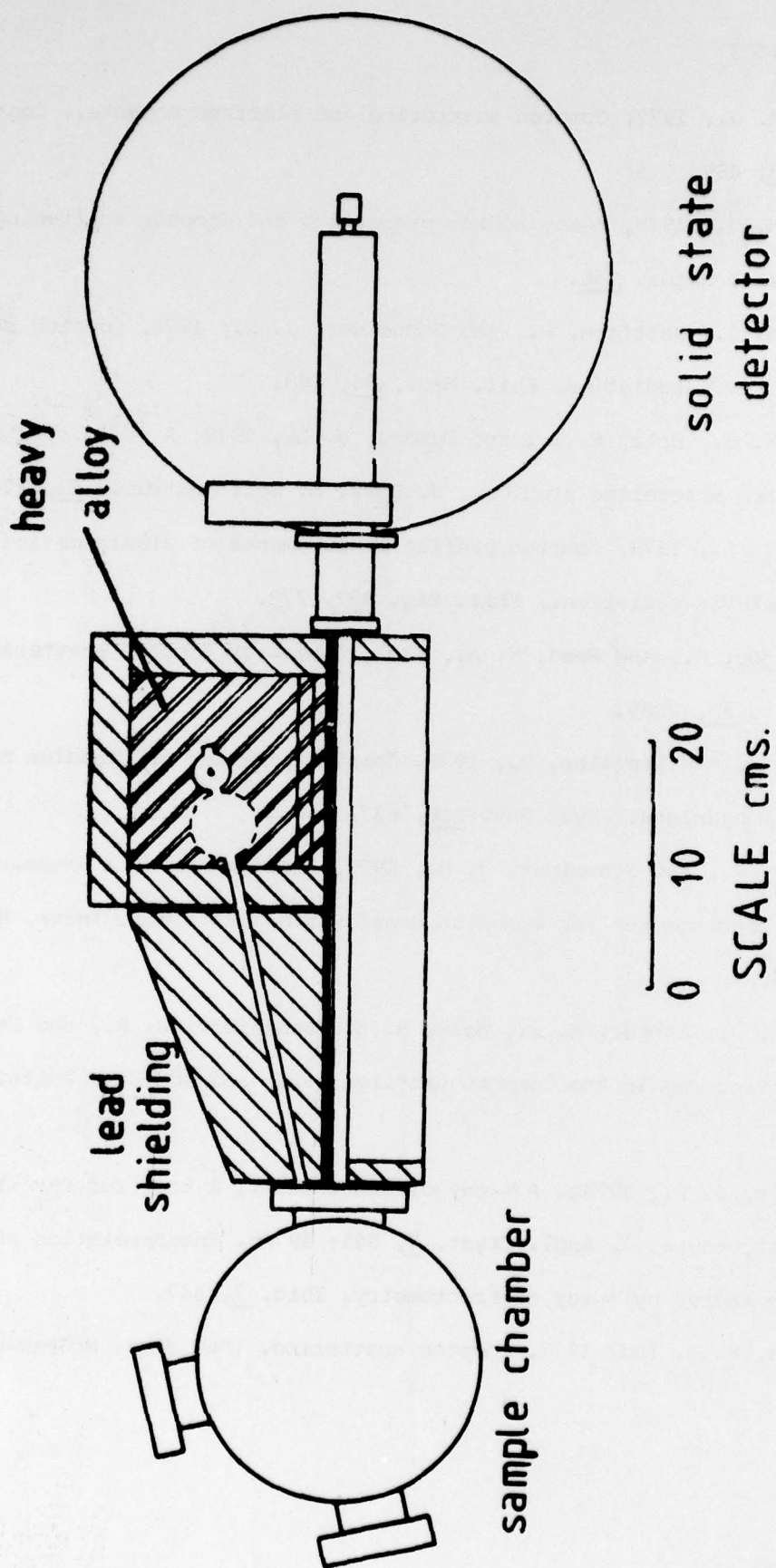
SHIELDING BLOCK FOR ^{198}Au SOURCE

pin

shutter

beam port

gold foil

lead

safety interlock

steel

SOURCE PIN DETAIL

ACTIVITY – (CURIES)

100

80

60

40

20

0

^{198}Au

^{199}Au

1

2

3

IRRADIATION – (DAYS) →
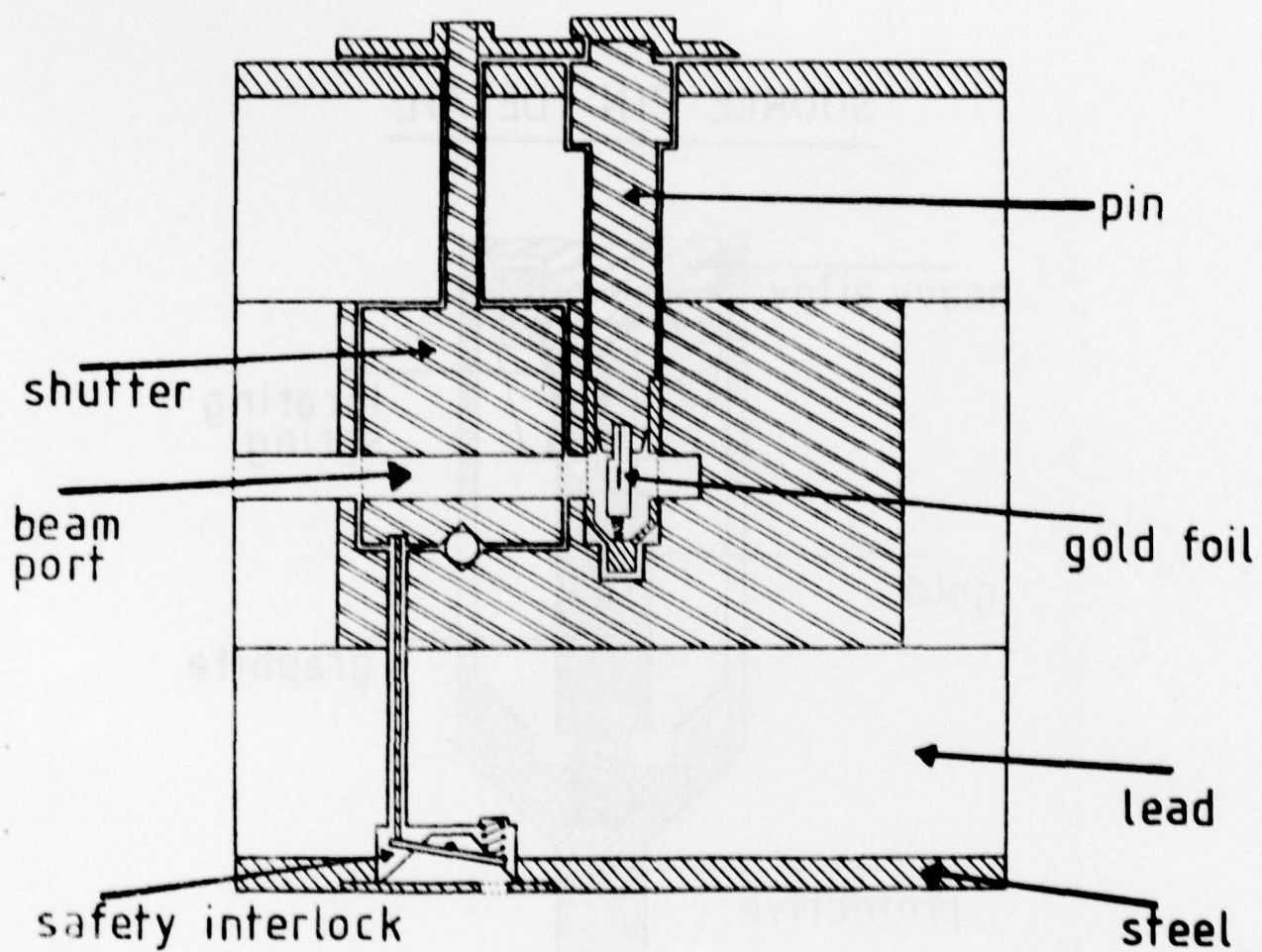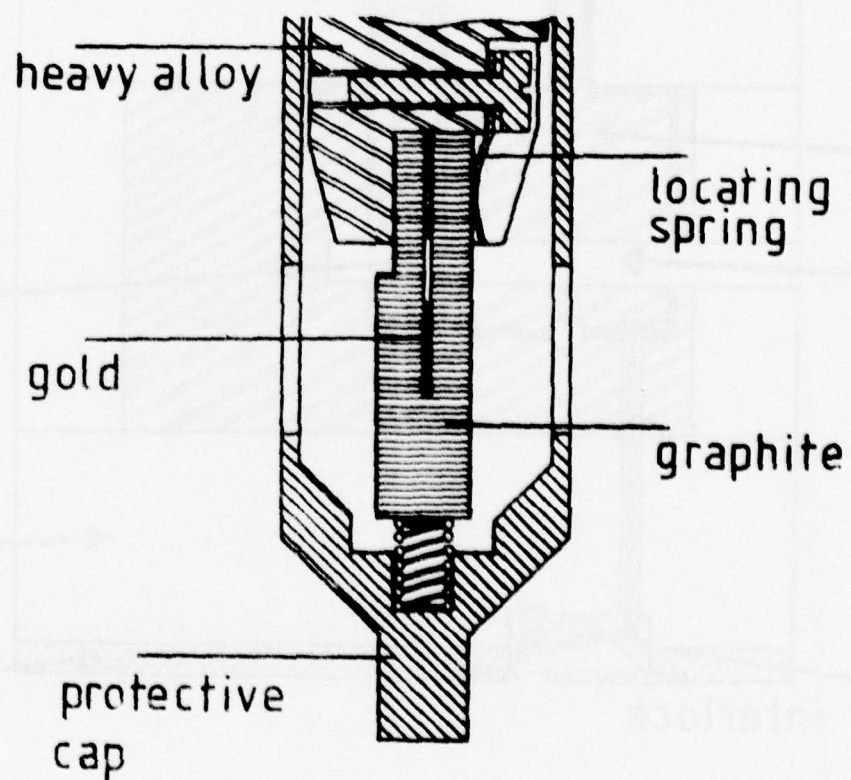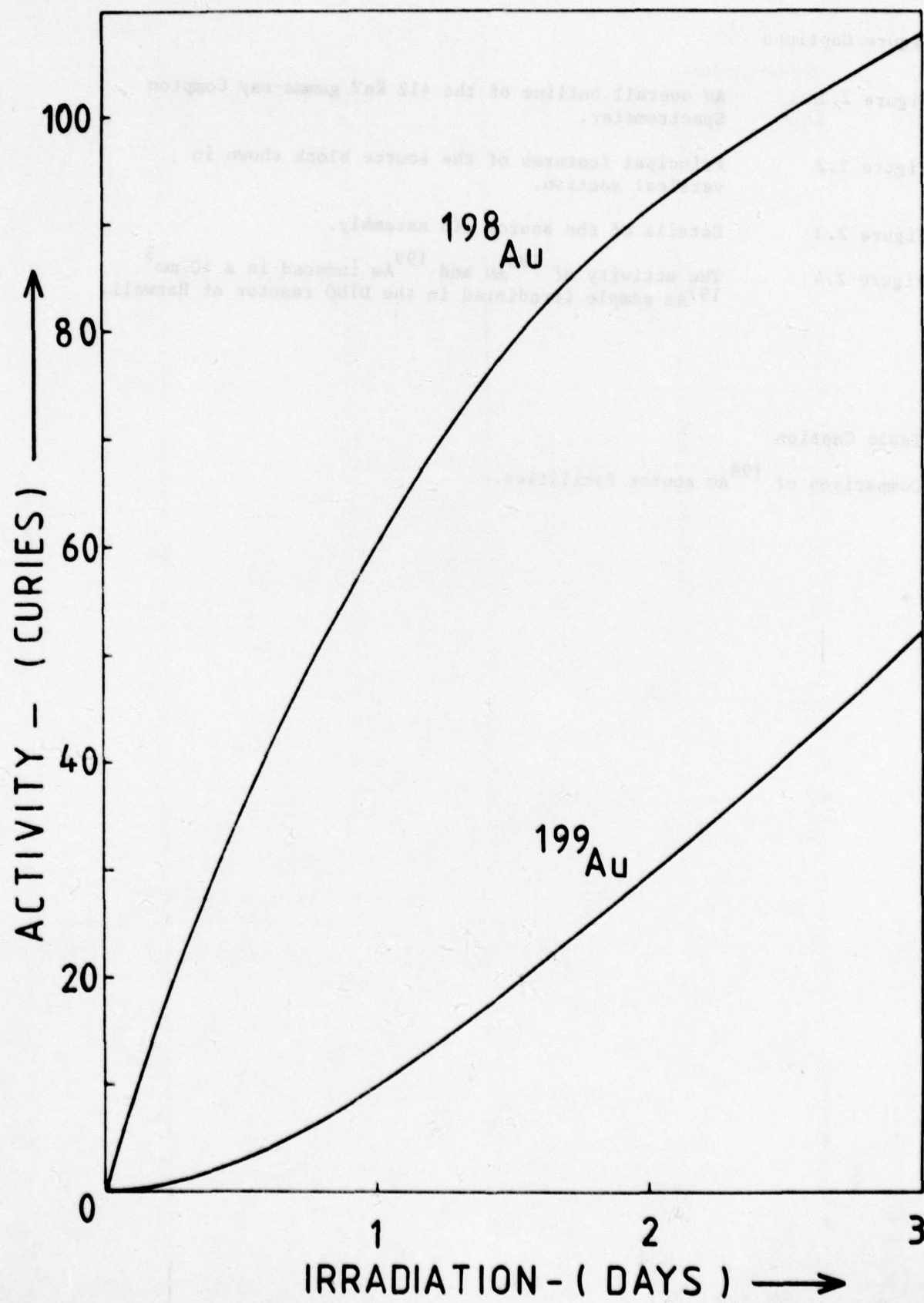
Figure Captions

Figure 2.1   An overall outline of the 412 KeV gamma-ray Compton
              Spectrometer.

Figure 2.2   Principal features of the source block shown in
              vertical section.

Figure 2.3   Details of the source pin assembly.

Figure 2.4   The activity of ^{198}Au and ^{199}Au induced in a 10 mm^3
              ^{197}Au sample irradiated in the DIDO reactor at Harwell.

Table Caption

Comparison of ^{198}Au source facilities.

SECTION 3: THE COMPTON PROFILE OF VANADIUM

§ INTRODUCTION

Preliminary measurements of the Compton profiles of (100), (110) and (111) slices of vanadium carried out at the Rutherford Laboratory were thwarted by the hardware and software faults on the PDP/8 computer (see section 2), and it was decided to make the measurements on the 412 KeV Compton spectrometer at the Hahn-Meitner Institute with the kind cooperation of Dr. Schneider and Dr. Pattison.

Vanadium was chosen for two reasons. Firstly the material itself is interesting because previous Compton measurements (at lower resolution) indicate that its momentum density is not as anisotropic as theory suggests. This is in sharp contrast with the behaviour of the next metal in the same group i.e. niobium, where good agreement between experiment and theory has been reported by the investigator (Pattison, P., Cooper, M., Holt, R., Schneider, J. R., Stump, N. (1977) Zeits. Phys. B27, 205). Secondly, as in the case of niobium, there is considerable interest in the behaviour of hydrogen in this metal and measurements on the hydride are also planned. The results obtained to date only represent an interim state in this project, and they are therefore only reported briefly.

§ EXPERIMENTAL

The equipment which was used has been recently described by Pattison and Schneider (Nucl. Instrum. Meths. 158, p.145, 1979). Approximately 10^5 counts were accumulated in the peak channel of the Compton profile for each of the three orientations of vanadium. The data were processed to produce the Compton profile, a multiple scattering correction being made by a Monte Carlo routine.

§ RESULTS

The results are presented in the accompanying figure in terms of difference profiles between the three orientations. The general nature of the oscillations can, as in the case of niobium, be explained in terms of Fermi surface geometry, but the size of the undulations is dependent upon the nature of the conduction electron wavefunctions. The two models, which are used for comparison, are the LCAO calculation of Laurent, Wang, and Calloway (Phys. Rev. B17, 455 1978 - solid line), and the APW calculation of Wakoh, Kubo and Yamashita (J. Phys. Soc. Jap. 40, p.1043, 1976 - dashed line). The agreement between them is very good, but both theories, which have been folded with the instrumental resolution function before comparison with experiment, show a large systematic deviations from the observations, vis. the amplitude of the oscillations in the theoretical anisotropies is much too large. X-ray diffraction measurements of the charge anisotropy by the 'paired reflections' technique are also in dis-agreement with these theories, but paradoxically position annihilation angular correlation profiles show larger anisotropies than this Compton data and are in reasonable agreement with both calculations.

In view of this confused situation it has been decided that a full investigation of the charge density in vanadium will be carried out with all the relevant techniques as a project of the I.U.Cr. Commission on Charge, Spin, and Momentum Density.

FIGURE CAPTION

Figure 3.1.   Difference Compton profiles for single crystal vanadium. The experimental difference profiles are plotted (open circles) as a function of momentum in atomic units. The solid line depicts the self-consistent LCAO calculation of Laurent, Wang and Callaway and the dashed line refers to the APW calculation of Wakoh, Kubo and Yamashita. Both theoretical curves have been convoluted with the instrumental resolution function which is a Gaussian with full width at half maximum of 0.41 a.u.

Vanadium

110 - 100

$\Delta J(p_z)$ 0 p_z(au)

111 - 100

$\Delta J(p_z)$ 0 p_z(au)

111 - 110

$\Delta J(p_z)$ 0 p_z(au)
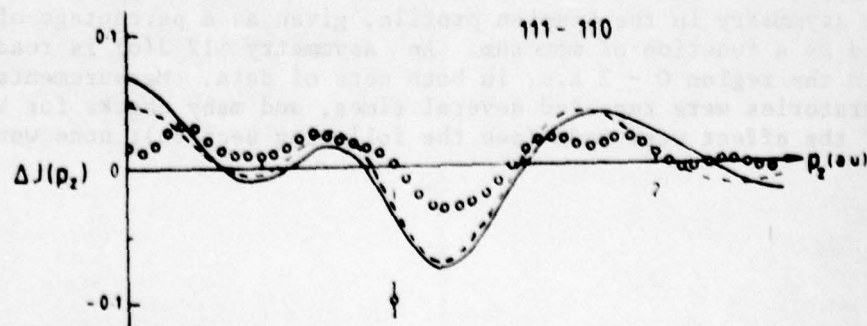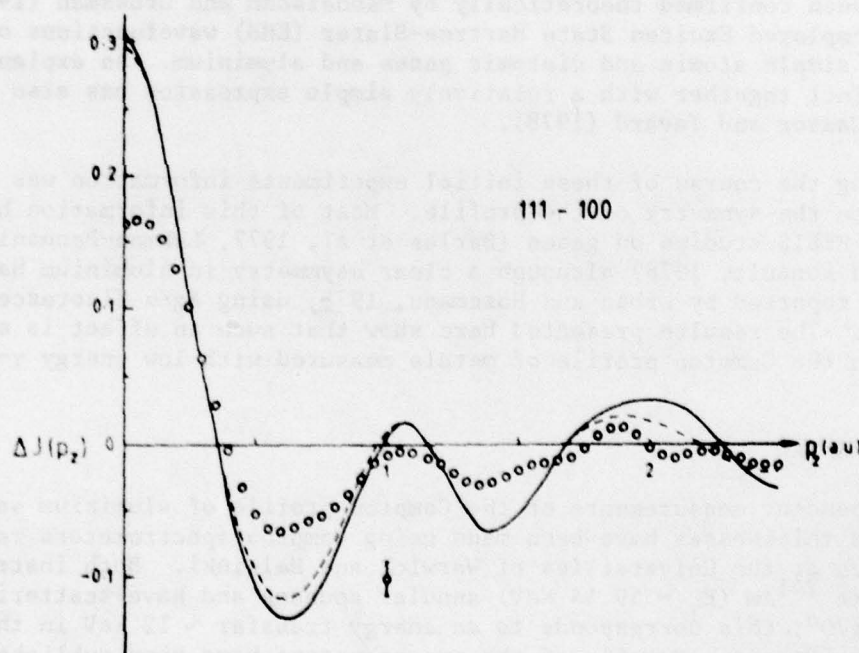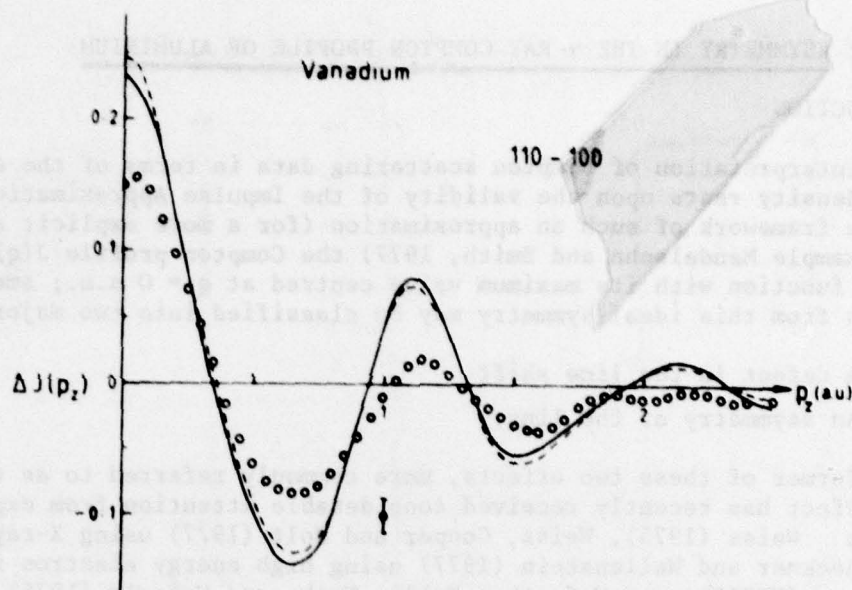
SECTION 4: ASYMMETRY IN THE γ-RAY COMPTON PROFILE OF ALUMINIUM

§ INTRODUCTION

The interpretation of Compton scattering data in terms of the electron momentum density rests upon the validity of the Impulse Approximation (IA). Within the framework of such an approximation (for a more explicit account see for example Mendelsohn and Smith, 1977) the Compton profile $J(q)$ is a symmetric function with its maximum value centred at $q = 0$ a.u.; small departures from this ideal symmetry may be classified into two major effects:-

(i) A defect in the line shift

(ii) An asymmetry of the line.

The former of these two effects, more commonly referred to as the Compton defect has recently received considerable attention from experimentalists. Weiss (1975), Weiss, Cooper and Holt (1977) using X-rays, Barlas, Rueckner and Wellenstein (1977) using high energy electron impact spectroscopy (HEEIS), and McCarthy, Noble, Ugabe and Weigold (1976) using (e, 2e) spectroscopic techniques, have reported both positive and negative defects in a variety of materials. The results of these measurements have recently been confirmed theoretically by Mendelsohn and Grossman (1978) who have employed Excited State Hartree-Slater (EHS) wavefunctions on a number of simple atomic and diatomic gases and aluminium. An explanation of the defect together with a relatively simple expression has also been given by Gasser and Tavard (1978).

During the course of these initial experiments information was also obtained on the symmetry of the profile. Most of this information has come from HEEIS studies on gases (Barlas et al, 1977, Lahman-Bennani, Duguet and Rouault, 1978) although a clear asymmetry in aluminium has also been reported by Urban and Hosemann, 1978, using AgKα fluorescent radiation. The results presented here show that such an effect is also present in the Compton profile of metals measured with low energy γ-ray sources.

§ EXPERIMENTAL

Independent measurements of the Compton profile of aluminium samples of various thicknesses have been made using Compton spectrometers recently constructed at the Universities of Warwick and Helsinki. Both instruments use 5 Curie ^{241}Am (E_γ = 59.54 KeV) annular sources and have scattering angles $\sim 170°$; this corresponds to an energy transfer ~ 12 KeV in the scattering process. Details of the spectrometers have been published elsewhere (Cooper, Holt, and DuBard, 1978, and Manninen and Paakkari, 1978). Both groups have used the standard data reduction procedures, as described by Williams, 1977. The results of the measurements are shown in the figure where the asymmetry in the Compton profile, given as a percentage of $J(o)$, is plotted as a function of momentum. An asymmetry $\sim 1\% J(o)$ is readily evident in the region 0 - 2 a.u. in both sets of data. Measurements at both laboratories were repeated several times, and many checks for spurious causes of the effect were made (see the following section); none were found.

In all cases there was a broad 'bulge' on the low energy side of the profile centred around 1 a.u., i.e. at an energy of 47.7 KeV. Lehman-Bennani et al. (1978) have characterised such asymmetries by the shifts, δq, of the midpoint of a chord drawn at half the maximum height of the profile. In that nomenclature the present data yeild $\delta q = + 0.04 \pm 0.01$ a.u., i.e. a shift towards larger energy transfers.

Such small effects have only now become measurable in γ-ray Compton scattering experiments because of the increased statistical accuracy achieved with the more intense sources. Nevertheless an effect of this kind could, in principle, arise from many causes. In order to establish that the asymmetry was not due to artefacts of the equipment, or systematic errors in the data processing, an exhaustive series of checks, lasting several months, were carried out by both groups and are described below.

§ POSSIBLE SOURCES OF ASYMMETRY

These fall into two categories, vis. those associated with the performance of the experiments, and those associated with the processing of the data: both potential sources of error have been considered.

1. Contaminent radiation from impurities in the source.

   There are two possibilities here, either the source contains additional γ-ray lines at the energy of the bulge (47.7 KeV), or it contains a line at an energy between 47.7 KeV and 59.54 KeV which is detected after being preferentially Compton scattered at some angle.

   A pin hole collimator was used to look at several parts of the source directly, the spectra showed no lines in the region of interest; in particular tungsten fluorescence, from the tungsten alloy backing on the source, was not significant.

2. Spurious scattering and X-ray fluorescence from the sample chamber.

   Both spectrometers are made from lead, with small amounts of tungsten alloy shielding at strategic points. However fluorescent scattering of K X-rays is too weak, and could not produce the observed asymmetry.

   The possibility of the effect arising from a double or triple scattering event involving the sample and the container was eliminated by using a special sample chamber one order of magnitude larger than the standard one. The results were unchanged.

3. Germanium K shell X-ray escape peak.

   Incomplete absorption of elastically scattered [241]Am γ-radiation in the germanium detector produces a very small secondary peak around 48 KeV, but this is on the wrong side of the peak and is, in any event, far too small to influence the observations.

4. Multiple Scattering in the Sample

Samples of various thicknesses were used, always with the same result. Monte Carlo calculations of the multiple scattering indicated this cause was ruled out. In the figure the Warwick results are shown without a multiple scattering correction and the Helsinki results have been corrected for double scattering.

5. Incorrect Resolution Function

It proved impossible to appreciably alter the profile symmetry at low momentum by using a range of credible asymmetric resolution functions.

6. Absorption corrections

Several alternative methods of calculating the absorption in the sample, and along the air paths were tried, but the different procedures produced no significant changes in symmetry.

7. The Compton cross-section.

A modification to the Ribberfors (1976) cross-section, as suggested by Felsteiner (private communication) merely produced an asymmetry in the profile at high momentum.

8. Location of the Origin of the Momentum Scale

Improper peak location manifests itself as a small adjustment to the position and magnitude of the bump. However the bump is readily observable in the raw data and it cannot be removed from the processed data by artificially altering the scattering angle.

9. Other checks

In addition to the elimination of the 'red herrings' described above, the experiments have been repeated on heavier metals (V, Mo) at Warwick with similar, but more marked results, while an experiment on lithium produced a symmetric profile. At Helsinki the experiments on aluminium have been repeated using a 900 mCi disc source and consequently a different experimental geometry; again the effect was found. On the other hand a detailed study of beryllium (Manninen and Suortti 1979) failed to reveal any asymmetry.

§ CONCLUSIONS

The investigations described above suggest that for aluminium the impulse approximation is not valid when ^{241}Am radiation is used. Although this conclusion is in line with the results of low energy X-ray studies (Urban and Hosemann, 1978) and HEEIS experiments, it is somewhat surprising to find a significant effect with such a light scatterer. Clearly studies of heavier elements with ^{241}Am sources will suffer progressively from systematic errors of this kind.
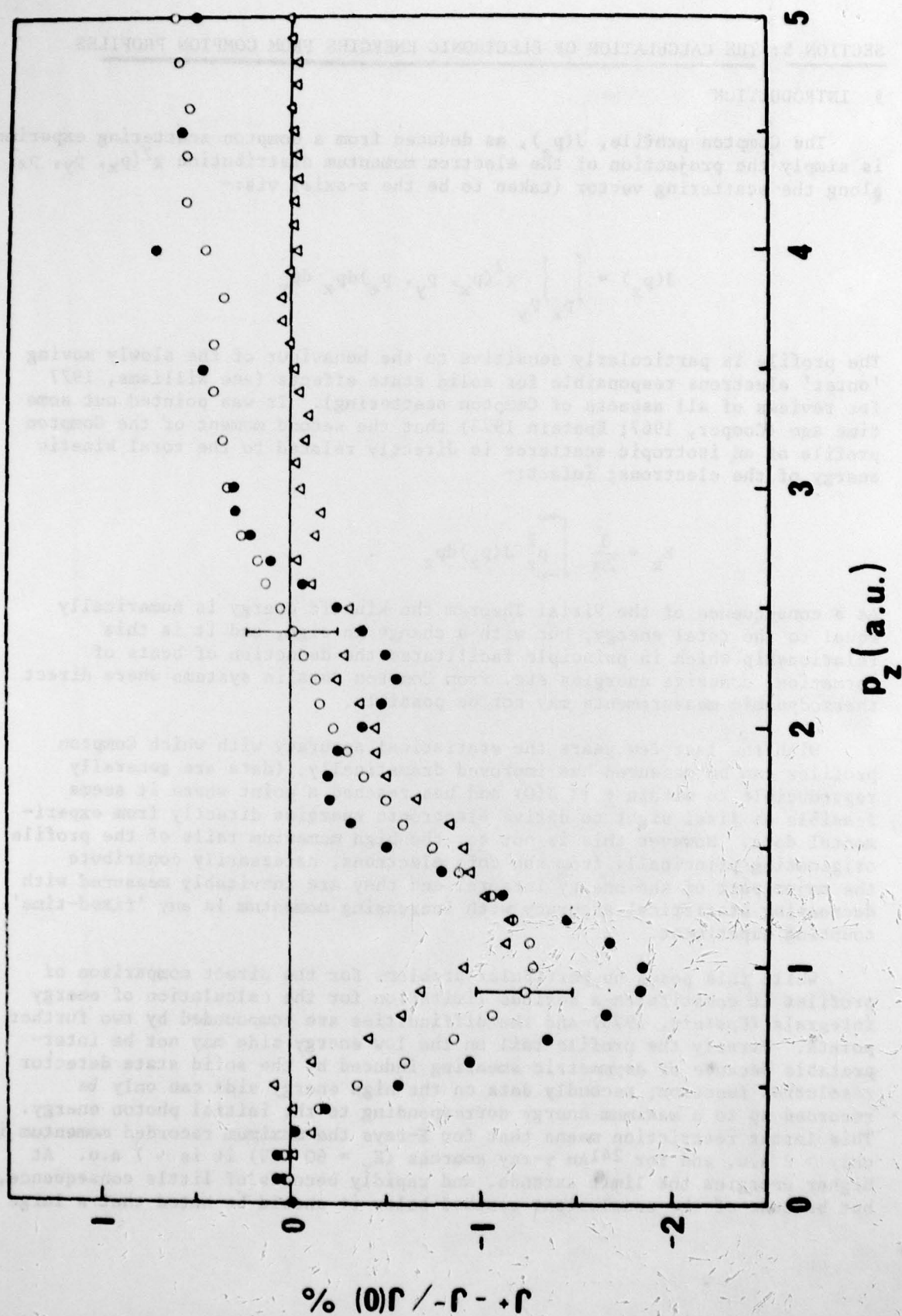
Any calculation of changes in the Compton profile resulting from deviations from the impulse approximation is difficult. Mendelsohn (private communication) has extended his EHS calculations to include the scattering of ^{241}Am γ-radiation from aluminium free atoms. However the calculation for the M-shell is limited to momenta below one atomic unit because of convergence problems, and this together with the solid state effects in the data renders a meaningful comparison difficult at present. The purpose of this paper is merely to establish the existence of an effect at this energy, further quantitative work is clearly called for.

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§ FIGURE CAPTION

Asymmetry in the Compton profile of aluminium. The graph shows the percentage asymmetry $\frac{J^+ - J^-}{J(o)}$ in the profile as a function of electron momentum, P_z, in atomic units. $J^+_{(P_z)}$ is the value of the profile on the high energy (i.e. low momentum transfer) side.

Key: Warwick results on a 0.6 mm thick sample Δ ; Helsinki results for sample thicknesses 0.25 mm O , and 0.025 mm $\bullet$.

p_z (a.u.)

% J+ - J- / J(0) %

SECTION 5: THE CALCULATION OF ELECTRONIC ENERGIES FROM COMPTON PROFILES

§ INTRODUCTION

The Compton profile, $J(p_z)$, as deduced from a Compton scattering experiment, is simply the projection of the electron momentum distribution $\chi^2(p_x, p_y, p_z)$ along the scattering vector (taken to be the z-axis) vis:-

$$J(p_z) = \int_{p_x} \int_{p_y} \chi^2(p_x, p_y, p_z) dp_x \, dp_y$$

The profile is particularly sensitive to the behaviour of the slowly moving 'outer' electrons responsible for solid state effects (see Williams, 1977 for reviews of all aspects of Compton scattering). It was pointed out some time ago (Cooper, 1967; Epstein 1973) that the second moment of the Compton profile of an isotropic scatterer is directly related to the total kinetic energy of the electrons; infact:-

$$E_k = \frac{3}{2m} \int_{-\infty}^{+\infty} p_z^2 \, J(p_z) dp_z \quad .$$

As a consequence of the Virial Theorem the kinetic energy is numerically equal to the total energy, but with a change in sign, and it is this relationship which in principle facilitates the deduction of heats of formation, cohesive energies etc. from Compton data in systems where direct thermodynamic measurements may not be possible.

With the last few years the statistical accuracy with which Compton profiles can be measured has improved dramatically, (data are generally reproducible to within $\pm \frac{1}{4}\%$ $J(0)$ and has reached a point where it seems feasible at first sight to derive electronic energies directly from experimental data. However this is not so: the high momentum tails of the profile originating principally from the core electrons, necessarily contribute the major part of the energy integral and they are inevitably measured with decreasing statistical accuracy with increasing momentum in any 'fixed-time' counting experiment.

While this poses no particular problems for the direct comparison of profiles it constitutes a serious limitation for the calculation of energy integrals (Epstein, 1973) and the difficulties are compounded by two further points. Firstly the profile tail on the low energy side may not be interpretable because of asymmetric smearing induced by the solid state detector resolution function; secondly data on the high energy side can only be recorded up to a maximum energy corresponding to the initial photon energy. This latter restriction means that for X-rays the maximum recorded momentum is only ~ 2 a.u. and for ^{241}Am γ-ray sources ($E_\gamma = 60$ KeV) it is ~ 7 a.u. At higher energies the limit extends, and rapidly becomes of little consequence, but because of the conclusions reached below it should be noted that a large

proportion of experimental work is currently carried out with ^{241}Am sources. In view of these points it is generally realised that the total energy cannot be deduced from an individual profile: however since it is thought that core electron distributions differ little between the free atom and condensed states, energy differences can be calculated from partial energy integrals of the form:-

$$E_{cohesive} = \int_{o}^{p_{max}} p^2 (J_s(p) - J_{fa}(p))dp$$

where the subscripts s and fa refer to solid and free-atom profiles respectively. The implicit assumption is that

$$\int_{p_{max}}^{\infty} p^2 (J_s(p) - J_{fa}(p))dp = 0 \quad .$$

This approach, if correct, has the merit that the choice of a small p_{max} minimises the statistical error of the energy integral (Epstein 1973, Nyanda and Williams 1979). Weiss (1978) for example has used $p_{max} = 1.5$ a.u. to calculate the cohesive energy of several metals from X-ray data. The work reported below questions the validity of using values of p_{max} generally below 10 a.u. - a limit which cannot be reached in low energy ($E_\gamma < 80$ KeV) Compton scattering experiments.

§ RESULTS

In order to assess the accuracy of partial energy differences it is necessary to study systems for which values have been established by other techniques. Thus attention has been restricted to simple molecules whose dissociation energies have been measured, and metals whose cohesive energies are known. In most cases available experimental data are limited to $p_{max} < 5$ a.u. and the majority of calculations have therefore been carried out on theoretical models where high momentum data are published. Representative results on the molecules H_2O and H_2, and metallic beryllium are presented below. Hydrogen is included to demonstrate that problems occur even in the lightest of materials.

However, before outlining each calculation one general point associated with interpolation should be mentioned. Compton profile data, including those tabulated for free atoms by Weiss, Harvey and Philips (1968), and Biggs, Mendelsohn and Mann (1975) are only quoted at widely-spaced intervals of momentum at high momentum: for example only three data points - 10.0, 15.0, 20.0 a.u. - are tabulated in the range 10 - 20 a.u. Consequently the energy integral, which must be computed using values interpolated on a fine mesh, depends upon the interpolation scheme which is chosen. Although these effects are small (< ½%) the binding energy itself is the small difference between two large integrals, and is therefore subject to greatly magnified errors (∿ 20%). Linear interpolation is clearly inadequate. Thus while cubic spline functions or parabolic interpolation can be cheerfully employed at low momentum, when they are used in the momentum range 5 - 20 a.u. the predicted energy difference does vary with the number and frequency of the data points.

In this study in order to circumvent the problem an analytic function of the form $J(p_z) = \dfrac{a}{(p_z^2 + \gamma^2)^3}$ was fitted at 10.0, 15.0, and 20.0 a.u., this is the form of the profile for a 1s electron described by a Slater-type orbital of the form $\psi(r) = Ae^{-\gamma r}$.

(a) The Dissociation Energy of the Water Molecule.

The near Hartree-Fock (NHF) model of Tanner and Epstein (1975) was chosen for this calculation because it has been found to be in good agreement with various sets of Compton data (Williams 1976). They quote a total energy of - 76.0630 a.u. per molecule (1 a.u. of energy = 27.2 eV) compared with the free atom value of - 75.8094 a.u., i.e. a binding energy of 6.9 eV. The dissociation energy calculated from the momentum wavefunctions, i.e. from $\langle p^2 \rangle$ is slightly different at 5.4 eV and it is this latter value which should be reproduced by the partial energy integral. [Both values are smaller than the observed dissociation energy of 10.08 eV, the difference arising presumably from the neglect of correlation effects].

The dissociation energy deduced from the Compton profile is shown in figure 1a, plotted as a function of p_{max}. Apart from a fortuitous correct value at $p_{max} = 1.0$ a.u., it can be seen that the correct value is only obtained if the integral is taken to $p_{max} = 10.0$ a.u., at which point the molecular and atomic profiles are indistinguishible. Furthermore if the calculation is terminated at 5 a.u. the dissociation energy is over-estimated by a factor to two.

(b) The Dissociation Energy of Molecular Hydrogen

In this case a theoretical Compton profile calculated by Smith, Thakkar, Henneker, Liu, Liu and Brown, 1977 from the configuration interaction wavefunction of Liu (1973) has been used. The Compton profile is averaged over a number of rotational and vibrational states as well as accounting for electron correlations, and is tabulated out as far as 5 a.u., above which point the profile is taken to be identical to that of the free atoms. For this, the lightest of molecules, it might be thought that the bonding effects are limited to very low momenta. However as far as the calculation of the dissociation energy is concerned the correct value 4.75 eV is only approached at relatively high momenta, as is readily evident in figure 1b.

(c) The Cohesvie Energy of Beryllium

For beryllium it has been possible to use experimental data obtained with ^{241}Am radiation by Manninen and Suorrti (1979) who quote data out as far as 10 a.u. (above which point equivalence with the free atom profile is assumed). The free atom calculation used in this instance is that of Benesch and Smith (1972) which includes configuration interaction. Using the same atomic calculation Manninen and Suorrti state that the cohesive energy deduced from their results is ~ 8 eV for $p_{max} > 2$ a.u. This is approximately true up to $p_{max} = 7$ a.u. but between 7 a.u. and 10 a.u. the cohesive energy approaches the observed value of 3.3 eV as is clear from figure 1c. Weiss (1978) also estimated the cohesive energy of beryllium

from X-ray data terminated at p_{max} = 1.5 a.u., and obtained a value of 14 eV. This is not inconsistent with figure 1c.

§ SUMMARY

Figures 1a, b and c are all similar in that they show that energy differences can only be deduced from Compton profiles if the data extends further than is generally thought necessary (i.e. up to 10 a.u.). In all cases investigated, apart from the lightest H_2, the momentum density in the region 5 - 10 a.u. differs sufficiently from the free atom value to make significant contributions to the energy integral. In the case of beryllium Weiss interpreted his results as indicating a contraction of the 1s momentum density, however an alternative explanation would be that the valence electron profiles have larger high momentum components than is generally supposed. Indeed the extent of the valence contribution to the Compton profile can be considerable, for example in fluorine (Snyder and Weber, 1978) the valence contribution at a momentum of 10 a.u. is some 10% of the total, although it is only 0.1% of J(0).

Finally in view of the problems associated with interpolation when energy values are to be deduced it is suggested that both theoretical and experimental profiles are tabulated at least at 1 a.u. intervals in the range 5 - 10 a.u. when they are published. Then it is possible, as has been shown above, to deduce energy differences with an accuracy $\sim$ 20% - 30%, but probably not better. While this may be of little practical use per se it can serve as a useful check on the accuracy of the representation in momentum space.

§ FIGURE CAPTION

The three graphs show dissociation energies, in eV, deduced from partial energy integrals, as a function of the upper limit p_{max} of the integral in atomic units of momenta. The vertical arrows in the two lower diagrams indicate the point above which free atom profiles are assumed. The theoretical values which the curves should approach are (a) 6.9 eV (b) 4.7 eV, and (c) 3.3 eV.

PARTIAL ENERGY INTEGRAL (electron volts)

P_{max}

(a) H_2O — 5.3 ev

(b) H_2 — 4.6 ev

(c) Be — 2.5 ev
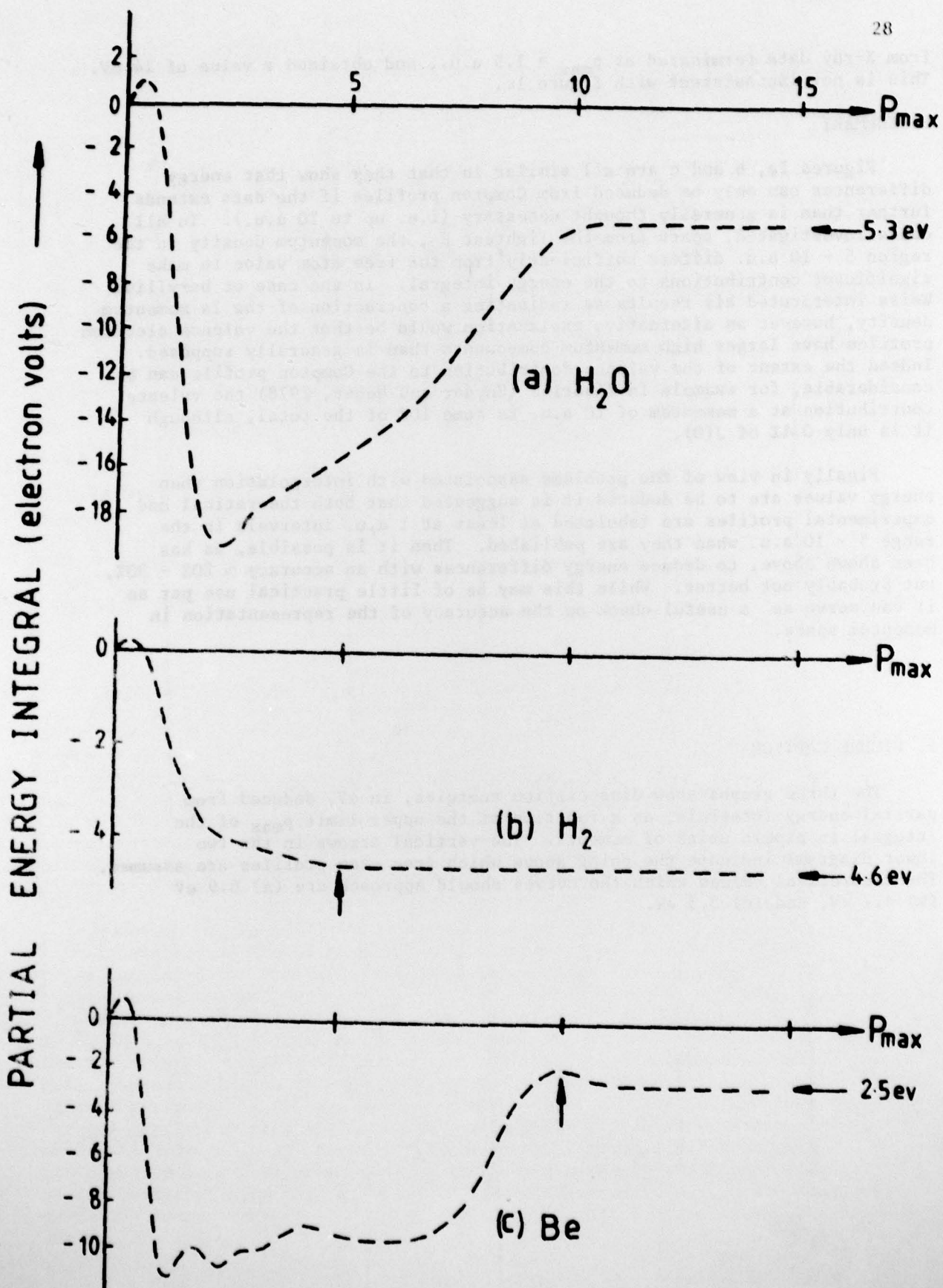
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SECTION 6:   SUMMARY

The primary purpose of this grant was to facilitate the involvement of Professor J. L. DuBard in the establishment of a 412 KeV Compton Spectrometer in the U.K. That objective was achieved, but apart from the measurements on vanadium the studies of metals and hydrides following from the groundwork have yet to be realised - primarily because of the failures of the data acquisition system and the deterioration of the detector. As noted earlier these faults have now been remedied and the planned programme of work will proceed.

This programme will include studies of metals and hydrides i.e. materials which have a strategic technological importance. Of the other studies included in this report the one which has most relevance to materials science is that described in section 5, namely the derivation of cohesive energies from Compton scattering data. In the case of materials which have low vapour pressures or high melting points (or which undergo a phase transformation before vapourisation) this technique may constitute the only non-destructive method of estimating cohesive energies.   Such a method may have a role to play in the case of such materials as TiB_2, TiC, and graphite which are of interest to research workers at AMMRC Watertown.

SECTION 7: ACKNOWLEDGEMENTS